Fatigue of high strength soda–lime glass: a constant stressing rate study using subthreshold indentation flaws

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Manuscript received 12 January 1982

A study is made of the dynamic fatigue properties of high strength (etched) soda–lime glass rods containing subthreshold Vickers indentation flaws, i.e. flaws with no visible signs of radial cracking. The fatigue characteristics of these flaws are distinctly different from those of corresponding post threshold flaws described in an earlier study: the strength level, the fatigue susceptibility, and the data scatter are all significantly higher. These results have special implications concerning the mechanical behaviour of ultra high strength fibres. In particular, the danger of extrapolating fracture mechanics data from macroscopic crack observations to the domain of the submicroscopic flaw is emphasised.

With the increasing demand for high strength glass components, particularly in the optical fibre industry, there is an associated need for a greater understanding of the nature of small scale flaws. This need is most evident in fatigue properties, where prolonged exposure to adverse environments under sustained stresses can reduce the strengths of initially pristine specimens to unacceptably low levels. The approach to the fatigue problem which has gained the widest acceptance in recent years is that of fracture mechanics, based on the hypothesis that some well defined microcrack grows to an unstable configuration in accordance with a specifiable crack velocity function. The laws of fracture mechanics, including the crack velocity function itself, are generally assumed to be determinable from the response of test pieces containing macroscopic cracks.

There is, however, some evidence from studies on high strength silicate glasses which appears to cast some doubt on the universality of conventional fracture mechanics. Metcalfe and co-workers[1,2] found that strength losses of carefully prepared E glass fibres were preceded by an incubation period in both static fatigue and ageing tests. They argued that this period must relate to a process of flaw initiation rather than propagation. In any case the estimated size of flaws necessary to account for fibre strengths near the theoretical limit is a few molecular spacings, at which level the concept of a well defined microcrack pre-existent in the glass hardly remains viable. This kind of consideration has led to the somewhat loose description of flaw types as either 'intrinsic' (characteristic of the glass structure itself) or 'extrinsic' (related to external factors such as handling history). Analyses of optical fibre fatigue data based strictly on the slow crack growth hypothesis appear to support this dichotomy;[3–5] the 'effective' exponent, n, in the commonly adopted power law form of the crack velocity function is found to be distinctly lower for 'pristine' than for 'damaged' surfaces; the value of this exponent is somewhat variable for the latter type of surface, but appears to become more representative of macroscopic crack behaviour as the severity of damage increases.

Recent work on glass specimens containing indentation flaws has provided some physical insight into the changing nature of flaws in different strength regions. The procedure basically involves the introduction of strength controlling centres by means of contact with a sharp, fixed profile indenter, using peak load as a test variable for adjusting the scale of the flaw. A preliminary study on borosilicate glass fibres[6] showed that post indentation inert strength varies systematically with load, but drops abruptly as the scale of the contact zone exceeds a critical value of a few μm. This drop corresponds to a threshold in the indentation process, above which well-defined radial cracks suddenly become apparent at the impression corners.[7] There is accordingly an inherent size effect where the fracture event which leads to failure undergoes a transition from initiation controlled to propagation controlled,[8] in keeping with the notions alluded to in the previous paragraph. Since the phenomenon of a fracture threshold is by no means unique to the indentation problem, requiring only that the stress field through which the crack evolves be highly localised (as around second phase or impurity centres),[9] there is a certain generality in the conclusions to be drawn from such observations.

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Extensive investigations of indentation flaws have been made during the past decade or so, but almost exclusively in the post threshold region.[10] The well developed radial crack system has a special appeal in the context of strength testing, both for its high degree of reproducibility and its amenability to direct microscopic observation (including observation during testing if necessary).[11] An important point which emerges from such investigations is the need to include a residual contact term in the crack driving force to failure, with consequent modification of the fracture mechanics under both inert[11,12] and fatigue[13,14] testing conditions. A comprehensive study of the dynamic fatigue (constant stressing rate) properties of soda–lime and borosilicate glasses by the present authors,[15] using a scheme which facilitates the incorporation of data taken at different loads onto a 'universal' plot for each material, confirmed the validity of macroscopically determined crack laws down to the fracture thresholds. It was noted as a point of discussion in this particular study that the procedure used might equally well be extended into the region of subthreshold loads, hopefully throwing some light on the intrinsic mechanisms responsible for degradation at ultra high strength levels. An additional experimental observation which added weight to this suggestion was the sudden drop in strength of soda–lime glass specimens indented below the threshold, down to levels characteristic of indentations containing radial cracks, when aged for a critical time in acid solutions;[16] no loss in strength was observed prior to radial crack 'pop-in', indicative of an initiation controlled incubation process analogous to that envisaged by Metcalfe and co-workers.[2] No such chemically enhanced pop-in was observed in borosilicate glass, however, this being attributed to the comparatively small residual contact driving forces about indentations in so called 'anomalous' glasses.[17]

Accordingly, a dynamic fatigue study in the subthreshold contact region forms the objective of the present paper. Our attention focuses on soda–lime glass, because of its strong tendency to the incubation behaviour just mentioned. The results of some preliminary tests on borosilicate will nevertheless be given brief mention in the discussion. To allow for ready comparison of results the experimental conditions used previously[15] are retained here as far as practicable. Thus, for instance, standard Vickers indenters are used to introduce the flaws, and the glass specimens are prepared in rod form. Although these conditions are somewhat restrictive in the range of flaw sizes that can be covered, the results obtained are expected to have broader implications, e.g. to high strength fibres. It will be demonstrated that the mechanics of fatigue immediately below and above threshold are markedly different in essential characteristics.

Experimental

Following the procedure described in Reference 15, rods 215 mm long and 5 mm diameter were cut from soda–lime glass cane.* These were annealed for 24 h at 520°C to remove any spurious surface stresses. An etch treatment was then administered in a solution of 10% $NH_4F.HF$/10% H_2SO_4 to nullify the stress concentrating power of preexisting surface flaws. Special care was taken with this last step, bearing in mind the intention to explore the high strength domain; the rods were accordingly mounted in a plastic holder which was agitated continuously in the acid bath, and were kept immersed for at least 10 min. After washing in methanol and drying, the glass surfaces were given a protective coating by dipping into varnish from both ends, leaving a central region $\approx$3 mm uncovered for subsequent indentation. Handling of the rods to this point was kept to a minimum, being confined to the portions near the ends. The specimens were stored in a dust free container.

For indentation the rods were mounted into V groove end supports fixed to the specimen stage of a Vickers microhardness tester. This simple expedient allowed for rapid alignment; as previously,[15] one impression diagonal of the pyramidal indentation was oriented perpendicular to the rod axis. The indentation surface was examined microscopically both before and after contact, before to preselect a site free of any unusually large etch markings and after to determine whether or not any cracking was evident. All indentations were made in air at a contact duration of 10 s. The threshold for radial cracking was $\approx$0·25 N, corresponding to an impression diameter $\approx$10 μm, but this load was subject to considerable scatter;[18] in this context small details in the conditions of testing, e.g. atmospheric humidity and imperfections in the geometry of the indenter (as evidenced by interchanging seemingly identical Vickers pyramids), appeared to exert a strong influence on the results. Our criterion for 'acceptance' of any given specimen was that there should be no visible radials at the indentation corners. The minimum working load specified for the microhardness testing equipment was 0·05 N, but it was generally found necessary to operate above this level to ensure that the indentation provided the dominant flaw (see below). Hence in comparison with the load range available in the previous study of the post threshold region (over 3 orders of magnitude)[15] the potential for using flaw size as a test variable is somewhat limited.

The accepted rods were then broken in four point flexure, using an inner span of 40 mm and an outer span of 90 mm.[15] The bending load was delivered by a crosshead testing machine, and was measured by either a conventional (strain gauge instrumented) or a piezoelectric load cell, according to whether the stress rate was less or greater than $\approx$100 MPa s^{-1}. Simple beam theory was used to calculate the stress at failure and the stress rate; linearity in the load–time response was maintained at all crosshead speeds, up to the highest strength levels reached, viz. $\approx$2 GPa. To assist

*Schott-Ruhrglas, GMBH. Wt% composition: $69SiO_2$, $13Na_2O$, 4CaO, $1B_2O_3$, $4Al_2O_3$, $2K_2O$, 4BaO, 2MgO, 1 other.

in orienting the system for maximum tension at the indentation a metal flag was attached to the end of each rod; it was then necessary only to ensure that the flag was kept vertical throughout the testing sequence. The indentation site was covered with a drop of either distilled water (for fatigue strength tests) or silicone oil (for inert tests) and subsequently covered with transparent tape immediately prior to bending. The taping served the dual purpose of minimising evaporation (a problem with the water at slower stress rates) and maintaining the specimens intact at failure (there being a greater tendency for untaped specimens to shatter at higher strengths); fulfilment of this latter requirement was essential to allow for determination of the failure origin, and thus for confirmation or otherwise of the indentation as the dominant flaw.

Results

Two features of the subthreshold flaw response were immediately apparent in the raw fatigue data from the tests in water: first, the strengths obtained were usually substantially higher than, and certainly never less than, those that would be expected from equivalent flaws with radial cracks; and second, the scatter in these strengths was relatively large. Figure 1, which shows the results obtained at two separate loads, illustrates these features. The Figure includes the baseline curve fit from Reference 15 to the previous post threshold results for soda–lime glass, plotted in terms of 95% probability bounds (±two standard deviations). In the data accumulation, distinction was made between those rods which broke at and away from the indentation flaws, and between those of the former kind which fell above and within the post threshold scatter band. Of these, only the clearly defined subthreshold indentation failures were retained. Certain reservations were held in rejecting the remaining failures, however: for the specimens which broke away from the indentations, most commonly at remnant etch markings in the vicinity of the inner supports, it is likely that data are being eliminated from the higher end of the strength spectrum; for the failures associated with indentation within the post threshold scatter band, where it can be argued that radial pop-in could have occurred during (or immediately prior to) flexure, there is a danger that we might be rejecting valid data at the opposite, lower end of the spectrum.

Notwithstanding these qualifications, the resultant data show some distinctive trends, which are evident in the reduced dynamic fatigue plot of Figure 2; here the error bars now represent one standard deviation about the mean strengths, in logarithmic coordinates, for 20 to 30 specimens at each stress rate. The associated smooth curve is a least squares linear fit to the strength data points from Figure 1, with data from

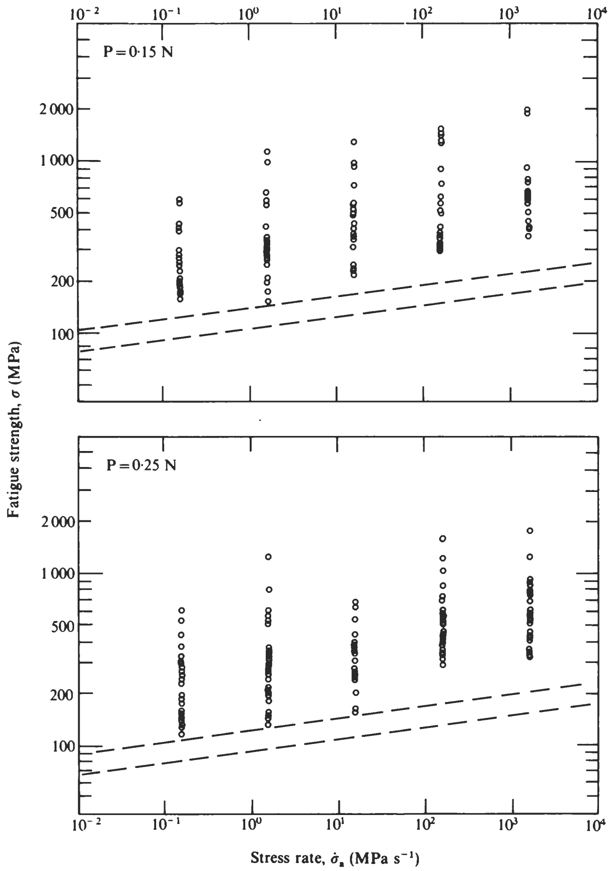

Figure 1. *Dynamic fatigue plot of σ (strength) against $\dot{\sigma}_a$ (stress rate), for soda–lime glass rods containing subthreshold Vickers indentation flaws tested in water. The baseline curve is the double standard deviation scatter band for post threshold flaws from Reference 15*

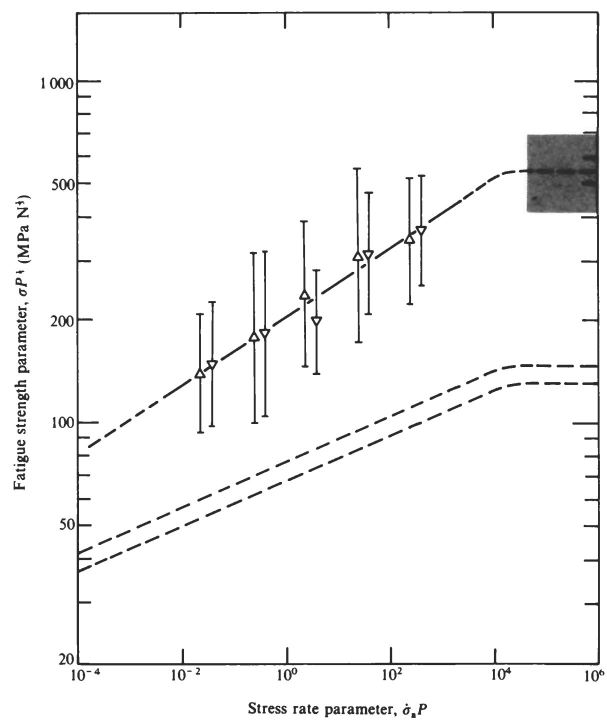

Figure 2. *Reduced dynamic fatigue plot for soda–lime glass rods containing subthreshold flaws tested in water. The upper shaded band is the inert strength plateau computed from data obtained in an oil environment. Coordinates incorporate indentation load,* P, *in a scheme for producing 'universal' curves. The baseline curve is the single standard deviation scatter band for post threshold flaws from Reference 15*

tests in the oil environment (20 specimens at the fastest available stressing rate of ≈ 1 GPa s^{-1}) determining the inert strength plateau. It will be noted that the coordinates of Figure 2 incorporate the indentation load in accordance with the scheme devised in Reference 15 for producing universal fatigue curves. This particular scheme has its foundation in the fracture mechanics of indentation flaws with well developed radial cracks, and as such has no strict justification for application to subthreshold behaviour. It is nevertheless used here as a convenient means of reducing data for comparison with the baseline post threshold curve. In any event the working range of contact conditions in the present study is hardly sufficient to allow for confirmation or otherwise of any predicted load dependence in the subthreshold domain.

Apart from the comparatively high strengths and large scatter already mentioned, the new results show one further trend of particular importance; the slope of the linear fit in the fatigue region is significantly greater than that of the post threshold curve. In terms of the fracture mechanics hypothesis this slope can be equated to the quantity $1/(n'+1)$, where n' is an 'apparent' exponent in the power law representation of the crack velocity function.[15] The mean value of n' is therefore distinctly lower for the subthreshold curve, $9{\cdot}0 \pm 0{\cdot}8$ as compared to $14{\cdot}0 \pm 0{\cdot}3$ (standard error bounds). For the post threshold exponent the theoretical analysis, which has to take explicit account of the residual contact driving force acting on the radial cracks, indicates that the 'true' value (i.e. representative of macroscopic crack response) is obtained via a simple multiplication factor of 1·31[19] (i.e. $n = 18{\cdot}4 \pm 0{\cdot}4$[15]). In the absence of an explicit fatigue model for flaws without radial cracks the basic form of any analogous 'transformation equation' for the subthreshold exponent cannot, of course, be specified.

Discussion

The results described in the previous section present clear evidence for a difference in the mechanics of fatigue on either side of the radial fracture threshold. This result has immediate implications concerning the use of fracture mechanics data to predict the lifetimes of ultra high strength components. In particular, we cannot extrapolate macroscopic crack characteristics to the domain of pristine glass surfaces without due regard to the specific micromechanics of very small flaws. In this context it may be argued that the commonly adopted practice of force fitting strength data on fibres in accordance with conventional fracture mechanics theory, i.e. assuming a well defined Griffith like microcrack driven exclusively by externally applied tensile forces, is without physical foundation. This is not to say, of course, that such fitting procedures are unjustified as a means of presenting data; it is simply that the parameters obtained from the fits are totally empirical, and as such strictly ought not to be used for predictions outside the data range. If, for example, a strength controlling subthreshold flaw were to pop in during service, as might occur on exposure to a suitably reactive environment (e.g. acidic as described earlier), the lifetime of a component could be orders of magnitude lower than that predicted.

There is an evident need to identify and understand the physical processes which lead to failure from subthreshold flaws. Unfortunately, unlike its post threshold counterpart where the offending radial cracks are clearly visible, this region is ill defined. The flaws are contained wholly within the deformation zone immediately surrounding the contact area and their size is accordingly of μm dimensions, a scale below that generally accessible by conventional optical microscopy. There is nevertheless some indication from section studies of larger scale indentations in soda–lime glass to suggest that radial cracking may have its origins in some precursor flow process beneath the penetrating indenter.[20–24] Hagan,[24] in particular, provides compelling evidence for a flaw controlled fracture threshold; shear 'faults' within the deformation zone initiate cracks at points of mutual intersection, where the stress concentrations are high. Taken to its logical conclusion, this picture implies that the subthreshold flaw does not have the character of a true microcrack, but rather that of a crack nucleus which requires a higher level of applied tension to produce failure instability. This would explain the difference in strength levels apparent for the two curves in Figure 2, and in the n' values deduced from the slopes of the curves. An initiation controlled mechanism of this type would also explain the incubation period for spontaneous loss of strength in soda–lime glass specimens aged in acid mentioned earlier in this paper.[16]

Thus the major point that emerges from this study is the demonstrable change in fatigue characteristics below the threshold for well defined radial cracking. It is as well to emphasise here that there are many issues left unresolved concerning the nature of very small flaws. For a start, we have considered an extremely limited range of indentation loads, immediately below the threshold, corresponding to average strength levels below 1 GPa. There is nothing to suggest that the fatigue response may not show further transitions at even smaller flaw sizes. We have already mentioned the distinction between intrinsic and extrinsic flaws which becomes apparent in the distributions of fibre strengths as the pristine surface state is approached. Second, we have reported on only one glass, soda–lime, whereas others, e.g. borosilicate, can show a widely different indentation response. The former is representative of 'normal' glasses, which deform by shear activated flow; the latter is representative of 'anomalous' glasses, where pressure-activated densification is dominant.[17] Our efforts to repeat the subthreshold fatigue test procedure on borosilicate glass have thus far met with limited success, due mainly to the relative difficulty of preparing rods of sufficiently high strength; attempts at removing handling flaws by etching served only to produce highly pitted surfaces, characteristic of glasses with separated phases.[25] Nevertheless, the preliminary data obtained on borosilicate specimens do show the same abrupt increase in strength below threshold, as would be expected from our initial studies on fibres of similar composition,[6] although the actual mechanisms of flaw evolution to failure could of course be quite different. In view of these considerations it would appear reasonable to advocate that future work concentrate on freshly drawn fibres rather than on rods, using specially constructed micro-indenters capable of delivering loads well below those available with standard hardness testing apparatus. Such experiments would be particularly instructive on a 'typical' anomalous glass (such as borosilicate), not only because they would fill a gap apparent in this study but also because of the universal usage of anomalous glasses for making optical fibres.

The ultimate aim of studies of the kind described here is a theoretical model of the rate limiting processes which control the mechanics of failure in ultra high strength materials. As intimated above, these processes may be quite different from those of ordinary crack growth, although the kinetic equations might bear a strong resemblance to those used in a fracture mechanics description. Any such model would need to take into account those essential forces which are responsible for both creating and driving the critical microcrack prior to and during stressing to failure. The variability in the subthreshold fatigue response (Figure 2), and in the actual threshold load, suggests that there might also be a need to incorporate certain probablistic elements into the description. Thus, while the latter concept is consistent with current statistical approaches to the strength behaviour of optical fibres, the underlying mechanical foundation upon which the statistics are based could require drastic revision.

Acknowledgements

The authors are grateful to P. L. Kelly and L. Respall for assistance with specimen preparation. Funding for the project was provided by the Australian Research Grants Committee.

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